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ON OÖLITES AND SPHERULITES[1]

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COLLOID-CHEMICAL INTERPRETATION OF ORIGIN OF OÖLITES

Schade's work.—The most important contribution toward a satisfactory interpretation of the origin of oölites and related structures was made by a member of the medical faculty of the University of Kiel, H. Schade, in 1909 and 1910, in his papers on the origin of urinary calculi and on the formation of concrements.[2] In these he demonstrated experimentally that concretionary bodies form when a substance passes from the state of an emulsion colloid (or "emulsoid") to that of a solid, and that if the change leads to the crystalline state the resulting structure is radial if the substance is pure; if, however, other substances, colloid or crystalloid, are precipitated along with it a concentric structure is developed. Corresponding with this law, natural holesterin gallstones, when 80 to 90 per cent pure, show a radial crystalline structure, while gallstones containing 25 per cent or less holesterin exhibit perfect concentric lamination.

Observations on iron chloride.—A substance which lends itself well to a demonstration of the process involved, because it requires no special preparation, is the commercial hydrated iron chloride. A concentrated solution of this salt, as was noted accidentally, exhibits all the characteristics of an emulsion colloid; for instance, a noticeable change of viscosity with change in temperature, stability in the presence of salts with polyvalent ions, etc. When dried at ordinary room temperatures, large spherocrystals are formed, perfectly spherical in shape when formed freely suspended in the heavy liquid, hemispherical in shape when formed in contact with the walls of the vessel or with the surface of the liquid. So far I did not succeed in preparing thin sections of these bodies, owing to the low melting-point of the salt (42° C.). In very thin layers, however, spread out on a slide, the growth is seen to take place in one plane only, yielding layers thin enough for optical study. This, combined with a macroscopic observation of the bases of the hemispheres, gave the following results:

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[1] A rock is called "oölitic," or an "oölite," if it contains or consists of small grains or units of dominantly concentric structure. A rock is called "spherulitic" (but not a spherulite) if it contains or consists of small grains or units of dominantly radial crystalline structure. An individual grain of a spherulitic rock is called a "spherulite." For an individual grain of an oölite the term "ovulite" might be used, which appears to be preferable to Kalkowsky's "oöid" not only for symmetry's sake. In this paper the term "spherite" will be used for all grains of the same origin irrespective of their structure.

[2] Heinrich Schade, "Zur Entstehung der Harnsteine und ähnlicher konzentrisch geschichteter Steine organischen und anorganischen Ursprungs," *Zeitschr. für Chemie und Industrie der Kolloide*, IV (1909), 175–80; "Ueber Konkrementbildungen beim Vorgang der tropfigen Entmischung von Emulsionskolloiden," *Kolloidchemische Beihefte*, I (1910), 375–90.

Spherites formed relatively rapidly during normal evaporation appeared structureless on cross-section with the center distinctly darker but not sharply separated from the rest. In other cases there was a sharply defined light center, and sometimes an outer lighter layer, distinct from the rest. Some spherites seemed to have formed through the union of several macroscopic drops, judging from an indistinct pattern shown in cross-section. When formed on a slide, growing in one plane only, they always exhibited a radial crystalline structure, the individual fibers being visible under the microscope. Superimposed on this radial structure there appeared in some of the spherites, on the same slide with the others, concentric lines outlining separate crystalline layers, which, however, all consisted of the same material.

Spherites formed during extremely slow evaporation (due to a crust formed on the liquid) showed a concentric structure of delicate layers of different color, especially in their outer part, representing in every respect true oölites. In several cases ten and more layers were counted in a radius of 2 mm.

These experiments illustrate well the principles of Schade: (1) the spherical shape of spherites is due to the tendency of the droplets, forming during the separation of the dispersed phase of an emulsoid, to coalesce;[1] (2) the difference between spherites of radial and concentric structure depends on the amount of other substance thrown out simultaneously with, and mechanically enmeshed in, the growing structure.

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[1] The solid crust which, in some experiments, formed on the viscous liquid containing the growing spherites was, in every case, ruptured upon further loss of water,

Ordinary concretions seem to differ only in size and, in many cases, in an excess of mechanically enmeshed materials. That these principles underlie the formation of most, if not all, sedimentary oölites, spherulites, and concretions is rendered probable by the fact that almost all substances which are known in one of these forms are also known in the others and are the same that are known to occur extensively in nature in the colloidal state. A brief review will emphasize this relation.

Review of natural oölites.—1. Silica: Des Cloizeaux found spherulites of clear silica in a jelly-like paste.[2] Spherulitic hyalite was observed by Jimbo from the Etchu province of Japan,[3] and similar hyalites with concentric structure from hot springs in the Ugo province were described by Takimoto.[4] Silica plays the rôle of the "binding" colloid in most pisolites and is an important constituent of most iron hydroxide oölites. Siliceous concretions are of wide occurrence. Silicic acid is the standard inorganic emulsoid sol of the laboratories.

2. Water: Among the hailstones we find all transitions between typical spherulites and spherites of concentric structure.[5] That

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and from the opening extensive *botryoidal surfaces* grew up, leaving cavities underneath the crust. Iron chloride, therefore, offers a complete analogy to all the common forms of natural gels, especially such as have a tendency to grow crystalline *in statu nascendi*, as, for instance, chalcedony. Cf. F. Cornu and H. Leitmeier, "Ueber, analoge Beziehungen zwischen den Mineralien der Opal-, Chalcedon-, der Stilpnosiderit-, Haematit- und Psilomelanreihe," *Zeitschr. für Chemie und Industrie der Kolloide*, IV (1909), 285–90.

[2] F. Roth, *Allgemeine und chemische Geologie*, I (1897), 591, Anm.

[3] K. Jimbo, "The Siliceous Oölite of Tateyama, Etchu Province," *Beitr. z. Min. Japans, Tokio*, 1905, pp. 11–75 (quoted from abstract in *Zeitschr. für Chemie und Industrie der Kolloide*, IV [1909], 287).

[4] T. Takimoto, "The Siliceous Oölite of Sankyo, Ugo Province," *Beitr. z. Min. Japans*, 2 (1906), 60–61 (quoted from *Neues Jahrb. für Min.*, etc., I [1907] 197).

[5] According to Schade the coarse layers overlapping onion fashion, frequently met with in hailstones, are not equivalent to the exceedingly delicate lamination of true oölites, but are due to plastic deformation under external pressure. See Schade, *Kolloidchemische Beihefte*, I (1910), 388; "Ueber die Koëxistenz des kristallinischen und kolloiden Zustandes," *op. cit.*, pp. 389 ff.

water vapor in the atmosphere frequently has the properties of an emulsion colloid is well known.

3. Hydroxides: The hydroxides of iron, manganese, and aluminum occur in all three forms with the exception of the last named, which is, as far as I know, not found in spherulitic or analogous structures. Aluminum hydroxide has, however, little chance to occur in the pure state. "Meta-"hydroxides in concentrated solution are known to be in the emulsoid state[1] and are widely distributed in that form in nature.

4. Phosphates: Phosphatic oölites are common in our western phosphate beds. Concretions are widespread and some show radial crystalline structure. The colloid origin of certain phosphates has long been recognized.[2] Because of the emulsoid character of their sols the phosphates are classed with the "gelatinous salts."[1]

5. Barite: Oölites consisting largely of barite with small amounts of gypsum and selenite were described by Wuestner[3] and Moore[4] from oil wells in Hardin County, Texas. They are of special interest because at least some of them undoubtedly formed in the wells after they were equipped, since they are found inside the tubing in sizes which could never have passed through the small mesh of the screen.[5] As the oil in the well has a temperature of 125° F. and contains free sulphuric acid, they offer a good example of an unquestionable case of an inorganic production of oölites. Fortunately some of these oölites, as shown on the micrographs of Wuestner's paper, Figs. 2–4, exhibit the same pattern of tubes radiating from the center which is so frequently seen in sections of sedimentary calcareous oölites[6] and which Kalkowsky, in his

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[1] Ostwald Wolfgang, *Handbook of Colloid-Chemistry*, translated by Martin H. Fischer (Philadelphia, 1915), p. 51.

[2] A. F. Rogers, "A Review of the Amorphous Minerals," *Jour. Geol.*, XXV (1917), 530–33.

[3] H. Wuestner, "Pisolitic Barite," *Jour. Cincinnati Soc. Nat. Hist.*, XX (1906), 245–50, 4 figs.

[4] E. S. Moore, "Oölitic and Pisolitic Barite from the Saratoga Oil Field, Texas," *Bull. Geol. Soc. America*, XXV (1914), 77–79.

[5] E. S. Moore, "Additional note on 'The Oölitic and Pisolitic Barite from the Saratoga Oil Field, Texas,'" *Science*, N.S., XLVI (1917), 342.

[6] For instance, in the oölites of the St. Louis limestone. They are also seen in two sections of bladderstones in the author's possession.

beautifully illustrated paper on the oölites of the Buntsandstein of northern Germany,[1] used as arguments in favor of an organic origin. The formation of the (unstable) barium sulphate sol in this case is possibly analogous to its formation in glycerin, as described by Recoura.[2] Barite concretions are locally found in shales.[3]

6. Calcium carbonate: In the sedimentary oölites we have all transitions from spherulitic to concentric structure. Perfect spheres of calcium carbonate, measuring 1 cm. and more in diameter, with excellent radial crystalline structure were described by the author from Miocene limestones of the Rhine valley.[4] Calcareous concretions are very common. In Drew's experiments, in which he proved the precipitation of calcium carbonate from sea water by *Bacterium calcis*, the first turbidity appearing in his solutions was caused by particles of such fine grain that they could be centrifuged only with difficulty. This suggests a colloid state. In the same experiments small spherulites were formed.[5] Vaughan allowed bottles containing Bahaman shoal-water muds strained through bolting cloth of fine mesh to stand over three months, after which he found in them numerous oölite grains which had grown to such sizes as to preclude their passing through the mesh of the bolting cloth.[6] The gel of calcium carbonate resulting from the precipitation of calcium carbonate from a solution of water-soluble calcium salts and its tendency to form spherulites have long been known. Owing to the elaborate studies of Buetschli, calcium carbonate is perhaps the best known of the "gelatinous salts."[7]

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[1] E. Kalkowsky, "Oölith und Stromatolith im norddeutschen Buntsandstein," *Monatsber. d. Deutsch. geol. Ges.*, LX, Part I (1908), 68–125, especially p. 122.

[2] M. A. Recoura, "Sur le sulfate de baryum colloïdal," *Compt. Rend.*, CXLVI (1908), 1274–76.

[3] See, for instance, J. P. Rowe, "Nodular Barite and Selenite Crystals of Montana," *Am. Geologist*, XXXIII (1904), 198–99; for a case in which the primary origin of these concretions is obvious see W. H. Bucher, "Beitrag zur geologischen und palaeontologischen Kenntnis des jüngeren Tertiärs der Rheinpfalz," *Geognostische Jahreshefte*, XXVI (1913), 31.

[4] Bucher, *op. cit.*, p. 80.

[5] G. H. Drew, "On the Precipitation of Calcium Carbonate in the Sea by Marine Bacteria," etc., *Carnegie Publication No. 182* (Washington, 1914), pp. 30–31.

[6] T. W. Vaughan, "Preliminary Remarks on the Geology of the Bahamas," etc. *Carnegie Publication No. 182* (Washington, 1914), pp. 51–53.

[7] Q. Buetschli, *Abh. Goettinger Akad.*, N.S., IV, 1908.

7. Siderite: Siderite is occasionally found in spherulitic form in cavities of basalt (spherosiderite).[1] Dana, Naumann-Zirkel, and others mention the existence of oölitic siderite. The only description of such an occurrence that has come to my attention is that of a specimen described by Dewalque from the Belgian coal measures.[2] Sideritic concretions ("clay ironstone") are commonly associated with caustobioliths.

This association is significant, since a colloid form of siderite was described by Van Bemmelen from the upland bogs of the Dutch province Drenthe, where it is found in the form of concretionary masses in irregular distribution in the peat.[3]

North of Preston, in Bath County, Kentucky, the Devonian limestone, which normally is dolomitic and more or less cherty, is partly replaced by a siderite. A specimen of the ore, presented to me by Dr. A. M. Miller, shows numerous small oölitic grains (0.5 mm. in diameter) of a light green silicate, uniformly distributed through the dark brown rock, which, under the microscope, exhibit a delicate concentric structure, rather indistinct in some grains (Figs. 1 and 2). The surrounding groundmass offers the usual appearance of interlocking siderite crystals. But at the contact with the oölites the siderite crystals show the sharp outlines of perfect rhombohedrons extending into the body of the oölites without disturbing their concentric structure. In this remarkable case the silicate oölites apparently formed in free suspension in a matrix which must have been an amorphous mud or a gel of iron carbonate, which later crystallized out before the oölites had lost their gelatinous character.

The necessary reducing conditions under which this local deposit originated within the coral-bearing dolomites of the Onondaga Sea may have existed in a depression on the bottom of the shallow sea in which the water lay stagnant,[4] or in a lagoon, which would not seem improbable if the assumption of a large island or peninsula in the Onondaga Sea, just to the northwest of this locality, is correct.[1]

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[1] As, for instance, at Steinheim, Hessia, where the author had occasion to observe and collect it.

[2] G. Dewalque, *Ann. d. l. Soc. géol. d. Belg.*, XV, Bulletin (1888), p. lxxx.

[3] J. M. Van Bemmelen, *Zeitschr. für anorg. Chemie,* XXII (1899), 313.

[4] See, for instance, J. Murray and J. Hjort, *The Depths of the Ocean* (London, 1912), p. 257.

These observations render it probable that other sedimentary siderite deposits, especially such as are closely associated with caustobioliths (black shales, coal, etc.), first separated out in the colloidal form and subsequently assumed crystalline character.

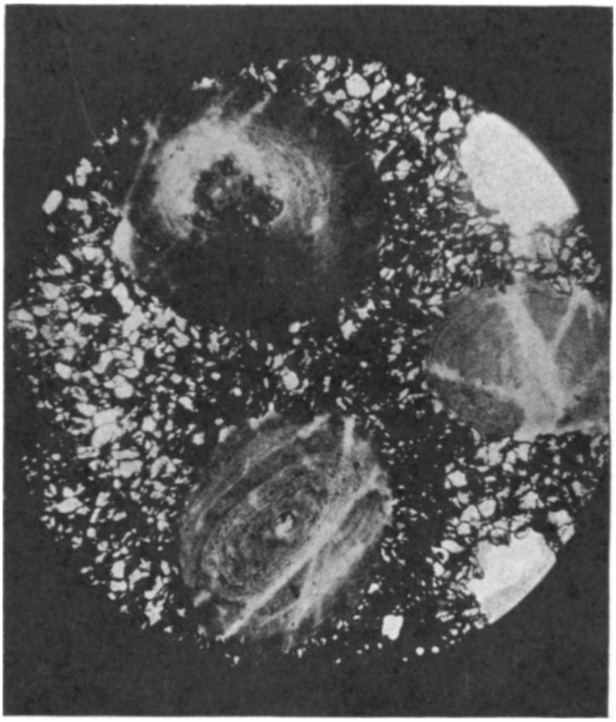

FIG. 1.—Photomicrograph of oölitic grains of a light green silicate in a groundmass of siderite and (secondary) iron hydroxide, forming a local facies of the Onondaga limestone near Preston, Bath County, Kentucky. Note the free edges of the siderite crystals extending into the oölitic grains, cutting through their structure, mostly without disturbing it; also the discordance between outline and structure of the grain near the lower margin. (The longest diameter of this grain measures 0.7 mm.)

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[1] C. R. Stauffer, "The Middle Devonian of Ohio," *Geol. Surv. of Ohio*, 4th Ser., Bulletin 10, 1909, Pl. 14; C. Schuchert, *A Text-Book of Geology*, Part II (New York, 1915), Pl. 15B; C. Butts, "Geology and Mineral Resources of Jefferson County, Kentucky," *Kentucky Geol. Surv.*, Ser. 4, Vol. III, Part II, Fig. 3.

8. Silicates: Some hydrous iron silicates, as, for instance, the greenalite of the Lake Superior region, and hydrous iron-aluminum silicates, as, for instance, the chamosite of the Jurassic "Minette" ores of Europe and the similar silicates of our Silurian ores, occur extensively in the oölitic form. They show all characteristics of original gels. In the case of greenalite the tendency to aggregate into numerous spherical grains was evident in the experiment.[1] Distinct oölitic structure is also seen in a specimen of fire clay, from the base of the Pottsville from an unknown locality in eastern Kentucky, which was presented to me by Dr. A. M. Miller. A similar occurrence was described by Dr. W. A. Tarr at the last meeting of the Geological Society of America. For the present the question must remain undecided if in these oölitic clays the binding substance is silica or a silicate.[2]

9. Iron disulphide: Only one case of an oölite has come to my attention in the formation of which pyrite seems to have had an independent part; that is, in which it is more than merely a mechanically enmeshed constituent or of secondary origin. In the upper Lias of Northwestern Germany there occur beds which consist partly of massive pyrite cementing the shells of belemnites and ammonites, and partly of nearly black limestone rich in pyrite, the two phases grading one into the other. In the black limestones oölites are found of a deep black color, measuring 0.5–3.0 mm. in diameter, and consisting of sharply defined alternating layers ("shells") of yellow pyrite and black calcium carbonate (soluble in cold HCl with effervescence).[3] Spherical nodules of pyrite with a fibrous radiated structure are not uncommonly found in shales,[4] and ordinary concretions are common. Recently a

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[1] C. R. Van Hise and C. K. Leith, "The Geology of the Lake Superior Region," *U.S. Geol. Surv. Mon.*, LII (1911), 522–25.

[2] Cf. Rogers, *op. cit.*, p. 535 ("imperfect pisolitic structure" in a clay consisting largely of the amorphous equivalent of crystalline kaolinite). A fire-clay containing small rounded bodies which are nearly pure alumina was described by Greaves-Walker in *Trans. Amer. Ceram. Soc.*, VIII, 297; quoted from Ries, "Clays" (New York, 1914), 52–53.

[3] T. Brandes, "Die faziellen Verhätnisse des Lias zwischen Harz und Egge Gebirge," etc., *N. Jahrb. für Min.*, etc., Beil. Bd., XXXIV (1912), 391.

[4] For a good example see, for instance, E. M. Nörregaard, *Meddelelser fra Dansk Geol. Fören*, XI (1906), 105.

black amorphous form of iron disulphide was described by Doss, who called it Melnikowite.[1] It is found in the form of small lenses and occasional thin incrustations of Pelecypod shells in gray clays of Miocene age and especially in solid layers of a pyritic sandstone. These consist of a mixture of pyrite and of this amorphous iron disulphide and contain abundant sand grains which, however, are everywhere seen to be suspended in the groundmass of disulphide. This must, obviously, have been in the state of an amorphous

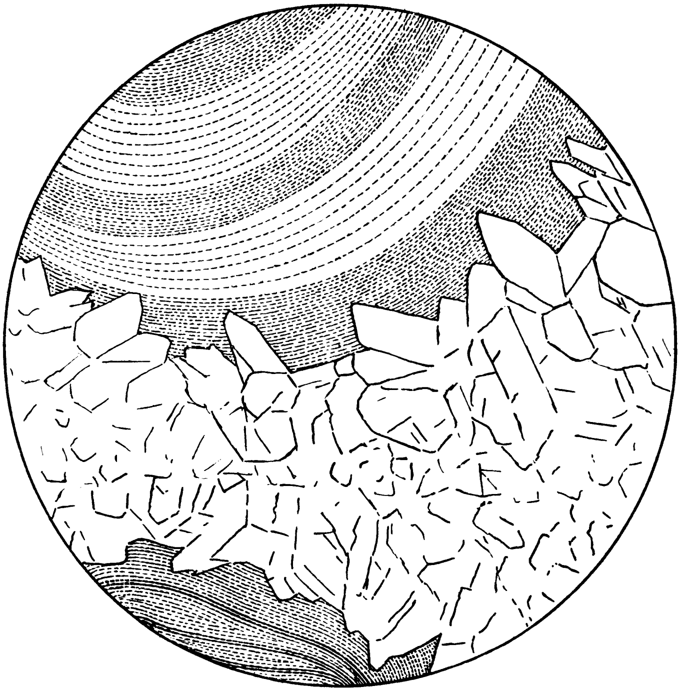

FIG. 2.—Sketch showing the free ends of the siderite crystals extending into oölitic grains. From the same slide as the preceding figure. The large, nearly perfect crystal at the contact in the lower right-hand quadrant measures 0.08 mm. in length.

NOTE.—Owing to the extreme softness of the silicate, A. C. McFarlan, one of my students, who prepared this and numerous other slides for me, was unable to avoid scratches on the oölitic grains. I am also under obligations to P. Scherrer for valuable assistance in the making of microphotographs.

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[1] B. Doss, "Ueber die Natur und Zusammensetzung des in miocaenen Tonen des Gouv. Samara auftretenden Schwefeleisens," *N. Jahrb. für Min.*, etc., Beil. Bd., XXXIII (1912), 662–713.

precipitate or of a gel at the time when the sand grains were dropped into it. The pyrite, which forms part of the layers, must therefore represent the final crystalline product of a series of transformations starting, according to Doss's interpretation, with the gel of hydrated iron monosulphide (FeSH.OH). This assumption is based on the experiments of Feld, who demonstrated that the iron monosulphide, which results from the action of hydrogen sulphide on iron salts, is changed within a few days into amorphous iron disulphide, when hydrogen sulphide is allowed to pass through it in the presence of free sulphur and a reducing agent[1] (conditions realized in the natural sapropels).

In these experiments the precipitate of the monosulphide was of black color and voluminous. In changing into the disulphide it turned brown and settled into a compact mass on the bottom of the vessel, sharply separated from the liquid above and undisturbed by the bubbles of hydrogen sulphide passing through it; it also formed a "mirror" of metallic brown color on the vertical walls of the vessel.[2]

This sudden change in the physical properties of the precipitate of the monoxide, the sol of which is a typical suspensoid, certainly justifies the suspicion that the iron disulphide formed a gel, passing through an emulsoid state.

It should be emphasized that any one of the substances referred to above may be present in an oölitic grain as the binding colloid or as an accidentally enmeshed crystalloid. The latter case is illustrated by many of the numerous organic oölitic structures, like animal and vegetal pearls, gallstones, urinary calculi, and many other similar bodies occasionally found in the tissues of the animal body, in which the percentage of organic substance, in those cases the binding colloid, is often very small.

SIGNIFICANCE OF THIS INTERPRETATION

This brief survey justifies the assumption that most, if not all, oölitic and spherulitic grains were formed by at least one constituent substance changing from the emulsoid state to that of a solid; that the spherical shape of the grains is due to the tendency of the droplets forming during this process of separation to coalesce; and that the difference between radial and concentric structure depends on the amount of other substance thrown out simultaneously with and mechanically enmeshed in the growing structure.

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[1] W. Feld, *Zeitschr. für angew. Chemie*, XXIV (1911), 97–103.

[2] Feld, *op. cit.*, p. 101.

The chief value, at present, of this interpretation of the origin of oölites to the geologist lies in the fact that it gives a new direction to future work. The questions to be answered in each case are:

1. What factors determined the colloid dispersion of the salt and what was the medium of dispersion?
2. What caused the separation of the colloid from the dispersion medium?
3. What made possible the suspension of the growing spherite?

The chemical geologist will have to decide in each case whether the factors involved in the first two questions were physical (for instance, the presence of protective colloids) or chemical (for instance, the nature and quantity of other substances in solution) or biological (for instance, the action of bacteria).

INTERPRETATION APPLIED TO SEDIMENTARY OÖLITES

Shape of grains due to growth in suspension.—The last question, on the other hand, is of special interest to the stratigrapher. We were accustomed to think that calcareous or limonitic oölites owe their spherical shape to constant rolling on the sea (or lake) bottom. While it cannot be said that such an origin is impossible, there is experimental evidence which raises doubts in my mind as to whether agitation of the suspension or dispersion medium would ever allow of the formation of such structures. Besides, there are now very few cases left in which such an interpretation might seem necessary.

The alternating layers of silica and of carbonate of the pisolites form while the spherites are being carried up by the current of the flowing spring water. The layers of hailstones form as they fall. Limonitic oölites form in the gels of organic iron salts on the bottom of Swedish and Finnish lakes.[1] Oölites of bauxite and of limonite form in residual clays.[1] The calcareous oölites of Great Salt Lake form suspended in the jelly-like masses of algae, as was described by Rothpletz.[2] Drew's artificial oölites formed in the agar-agar of his bacterial culture, and in Vaughan's experiments they grew in the soft amorphous calcareous muds which occur so abundantly on the shores of the Bahama Islands.

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[1] For references see F. Beyschlag, J. H. L. Vogt, and P. Krusch, *The Deposits of the Useful Minerals and Rocks*, translated by S. J. Truscott (London, 1916), II, 982.

Apparent exceptions.—I know of only two cases which at first sight at least seem to form exceptions to the rule that oölites grow in free suspension.

1. Gaub, in a splendid paper, described oölites containing numerous microscopic shells of a Foraminifer classed with the Miliolidae (*Ophthalmidium ooliticum*).[3] These he considers to have been incrusting forms which attached themselves to small fragments of shells, crinoid stems, etc. (now forming the nucleus of the oölitic grains), and, being rolled about in the amorphous calcareous mud, held it mechanically and perhaps even localized its precipitation. The fact that a very similar, not incrusting, species of the same genus *Ophthalmidium* is very abundant in the same layers suggests the possibility that these minute shells were only mechanically inclosed in the growing oölite and that surface tension may be responsible for their tangential arrangement. Schade observed, for instance, that in some pearl-like gallstones the holesterin crystals were all arranged tangentially, enmeshed in the binding colloid.[4] Some of the shells within the oölitic grains are flattened on the side facing the center of the oölitic grain. Others, however, exhibit similar deficiencies on the outer side. Gaub interpreted the former as evidence of attachment, the latter as evidence of mechanical wear. They may, however, both be due to a small amount of solution during the growth of the oölite.

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[1] C. K. Leith and W. J. Mead, *Metamorphic Geology* (New York, 1915), pp. 35–37; for literature on European "bean ores" see Beyschlag, Vogt, and Krusch, *op. cit.*, p. 990.

[2] A. Rothpletz, "On the Formation of Oölite," *Amer. Geologist*, X (1892), 279–82 (translated from *Botanisches Centralblatt*, LI [1892], 265–68).

[3] F. Gaub, "Die jurassischen Oolite der Schwaebischen Alb," *Geol. und palaeont. Abh.*, N.S., IX (1910), Heft 1. An earlier shorter paper, *N. Jahrb. für Min.*, etc., Part II, (1908), pp. 87–96, Pls. 7–8.

[4] Schade, *Kolloidchemische Beihefte*, I (1910) 385.

Since, at present, the original papers are inaccessible to me I cannot judge the degree of probability of this suggestion. It is, of course, possible that frequent interruptions in the normal process of growth of the oölitic grains, caused by a stirring of the calcareous muds during storms, permitted the *Ophthalmidia* to attach themselves to the grains and thereby to participate in their growth.

2. The second case comprises oölites in the formation of which filamentous algae are supposed to have had an active part. Such oölites were, for instance, recently described by Van Tuyl from the Ordovician of Iowa.[1] They contain in abundance "minute sinuous fibers similar to those which characterize the Girvanella type of calcareous algae." In oölites from the shores of the Red Sea, Rothpletz found "peculiar vermiform, and not rarely dichotomously branching, canals that are filled up with calcite."[2] In fresh-water springs and pools threadlike Schizophyceae are usually found associated in great numbers with the primitive types which cause the separation of the lime carbonate. Rothpletz, therefore, considered the vermiform structures of the oölites as "threadlike algae which were, of course, not themselves immediately concerned in the oölite formation, but by the company in which they lived were imprisoned with it." Since Van Tuyl states expressly that his oölites showed in addition to the supposed calcareous algae "good concentric and radial structure," there can be little doubt but that Rothpletz' interpretation may be applied to them directly. Similar canals might, however, also be produced by boring algae or fungi, the ramified canals of which are found permeating larger shells and pebbles as well as shells of Foraminifera in size comparable to the oölites.[3]

Relation of algae to oölites.—The rôle which Rothpletz assigned to such Schizophyceae as Glœocapsa and Glœothece in the formation of oölites appears to have been generally misunderstood. The cells of these primitive algae correspond in size to the largei forms of bacteria.[1] Bodies resulting from the precipitation of lime on or in the thick, jelly-like membrane surrounding the minute cells would therefore have a diameter fifty to a hundred times smaller than that of the oölites. Such bodies were, in fact, observed by Rothpletz in the oölites from Great Salt Lake as well as in those from the Red Sea, and were found to be mechanically incased in the oölites like the filamentous algae. "In quite delicate sections the calcareous substance [of the oölites] . . . . is interrupted by scattered, minute granules. If we dissolve the section cautiously and slowly in very dilute acid, the granules remain behind exactly in their original position, and we recognize in them the dead and crumpled Glœocapsa cells" (p. 280).

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[1] F. M. Van Tuyl, *Science*, N.S., XLIII (1916), 171; *Jour. Geol.*, XXIV (1916), 792–97.

[2] Rothpletz, *Am. Geologist*, X, 280.

[3] J. E. Duerden, "Boring Algae as Agents in the Disintegration of Corals," *Bull. Amer. Mus. Nat. Hist.*, XVI (1902), 323–24. They were also described from Ordovician Foraminifera and from numerous Siluric fossils (see F. B. Loomis, "Siluric Fungi from Western New York," *Bull. N.Y. State Mus.*, VIII (1900), No. 39, 223–26, especially Fig. 3 on Pl. 16).

From this it follows that if the Schizophyceae have at all an active part in the formation of oölites[2] it must be similar to that played by the closely related bacteria which separate calcium carbonate in the form of an emulsoid sol from solutions of calcium salts and thereby create conditions favorable for the growth of oölitic grains.[3]

The papers in which Wethered described Girvanella from many oölitic rocks of various ages[4] are, unfortunately, not accessible to me. The structure of certain calcareous oölites described by him, according to Rothpletz, seems "to have great resemblance to that of the Sinai oölite and is, perhaps, to be explained in the same manner." Others, however, judging from one of his figures reproduced in Harker's *Petrology for Students*,[5] represent true incrustations of organic fragments by the interlacing tubes of Girvanella and should therefore not be called oölites at all.[1] When exposed, true oölites may, of course, be incrusted in the same way.

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[1] Glœocapsa: 2 μ in diameter; Glœothece: 4–5 μ long; Anthrax bacillus: 6 μ long.

[2] Weighty reasons against this assumption were adduced by T. C. Brown, "Origin of Oölites and the Oölitic Texture in Rocks," *Bull. Geol. Soc. America*, XXV (1914), 754–57.

[3] As this process is probably due to a reaction of the calcium salts of the water with an alkaline excretion of the algae, we cannot, in this case, speak of "lime secreting" algae (cf. W. Pfeffer, *The Physiology of Plants*, I, translated by Ewart [Oxford, 1900], 133). Compare Drew's account of the action of denitrifying bacteria (*Carnegie Institution of Washington, Publication No. 182*, p. 30).

[4] E. Wethered, "On the Occurrence of the Genus Girvanella in Oölitic Rocks," etc., *Quar. Jour. Geol. Soc.*, XLVI (1890), 270–83; LI (1895), 196–206.

[5] Fourth ed., London, 1908, fig. 69, p. 261.

SOURCES OF ERROR IN INTERPRETING ORIGIN OF OÖLITES

Secondarily deposited oölites.—Erroneous conceptions concerning the origin of oölites may arise if no clear distinction is made between layers in which the oölitic grains are found *in situ* and such in which they were redeposited after transportation. The secondary origin of oölitic deposits may be recognized by the practical absence of a matrix, by the uniformity of size of grains (indicating sorting), by cross-bedding, or by the presence of substances accidentally enmeshed in the oölitic grains which are foreign to the surrounding matrix, etc. In such cases the oölitic grains were either washed by waves or currents from their place of origin and redeposited in water, or they were carried inland by the wind where they may have formed dunes, as they do now on the shores of the Red Sea and of Great Salt Lake.[2] The recognition of such secondary oölites and their correlation with synchronous primary deposits may, under some circumstances, convey valuable information to the paleogeographer.

It is often quite difficult to prove satisfactorily the origin *in situ* of an oölite. For this we must, in many cases, rely entirely on a microscopic study of the relation existing between the matrix and the oölitic grains. In some cases, however, the characteristic incrustations and massive growths, called "stromatoliths" by Kalkowsky,[3] are found associated with the oölitic grains and by their presence prove the primary nature of the oölites, as, for instance, in the Upper Triassic (Rhaetic) of England, the Mississippian of Belgium, the Lower Bunter of Northwestern Germany, the Tertiary of the Rhine Valley, etc.[4]

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[1] The term "pseudoöolite" has been used for grains imitating oölites; for instance, minute pellets of dense limestone in a limestone matrix (cf. O. M. Reis, *Geognostische Jahreshefte*, XXII [1909], 228).

[2] J. Walther, *Lithogenesis der Gegenwart* (Jena, 1894), pp. 659, 699, 849; A. W. Grabau, *Principles of Stratigraphy* (New York, 1913), pp. 468, 472.

[3] Kalkowsky, *Monatsber. Deutsch. geol. Ges.*, LX, I (1908), 68–125.

[4] Cf. O. Reis, "Ueber Stromatolith und Oolith," *N. Jahrb. für Min.*, etc. (1908) Part II, pp. 114–38.

Under the microscope these stromatoliths exhibit a structure of delicate layers identical with that of the oölites, with which they are often connected by all stages of transition.[1] They differ greatly, both in structure and in origin, from the coarse calcareous crusts which are formed by thick, felted masses of fresh-water algae and mosses on shells and pebbles.[2] The two can easily be distinguished when found associated in the same formation, as, for instance, in the Miocene limestones of the Rhine valley.[3]

The stromatoliths are the sedimentary equivalent of the calcareous and siliceous "sinter" of the hot springs; stromatolithic crusts of limonite are commonly found in lake and bog ores.

In the simple experiments with iron chloride, described in the first part of this paper, similar growths were observed in the same cases in which spherites with oölitic structure were produced. In these experiments flat expansions formed below the massive crust which sealed the liquid, and, to a smaller degree, also at the bottom of the vessel, corresponding in thickness to the radius of the oölitic grains and showing the same delicate concentric structure. There can be little doubt but that they represent the experimental reproduction of stromatoliths.

Subsequent alteration of oölites.—Another, more fundamental, source of error in the interpretation of the origin of oölites has been the more or less altered condition of most fossil oölites. Many calcareous oölites, for instance, have suffered complete recrystallization which, starting usually in the center of the grains,[4] replaces their original structure by a few crystals, or even a single large calcite individual, concentrating all impurities in a thin layer at their periphery. Such extreme cases have been interpreted as incrusted gas bubbles secondarily filled with calcite ("ent-oölites").[1]

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[1] Cf., e.g., Bucher, *Geogn. Jahresh.*, XXVI (1913), 78–79, and Pl. I, Fig. 22.

[2] Cf., e.g., J. M. Clarke, "The Water Biscuit of Squaw Island, Canandaigua Lake, N.Y.," *Bull. N.Y. State Mus.*, VIII (1900), No. 39, 195; W. Schmidle, "Postglaciale Ablagerungen im nordwestl. Bodenseegebiet," *N. Jahrb. für Min.*, etc. (1910), Part II, 105–22.

[3] Bucher, *op. cit.*, pp. 80–81.

[4] Cf. the illustrations and the discussion of the mechanism of this process in Reis, *Geogn. Jahresh.*, XXII (1909), 227–31, and Pl. 11, Figs. 25–30.

In other cases the substance of the oölitic grains has been completely changed, in some even before final deposition, in others during the diagenesis of the sediment, or still later through metasomatic or katamorphic processes, as in the dolomitic oölites,[2] some siliceous oölites,[3] and the great variety of oölitic iron ores, the interpretation of which, in most instances, is still far from being satisfactory.

[1] C. W. v. Gümbel, *N. Jahrb. für Min.*, etc., 1873, p. 302. The incrustation of air bubbles by calcium carbonate was observed by Knop in the hot springs at Nauheim. Cf. *ibid.*, 1874, p. 285.

[2] Cf., for instance, W. H. Sherzer and A. W. Grabau, "The Monroe Formation of Southern Michigan and Adjoining Regions," *Mich. Geol. and Biol. Surv.*, Publ. 2, 1910, pp. 35-37; T. C. Brown, "Origin of Oölites and the Oölitic Texture in Rocks," *Bull. Geol. Soc. America*, XXV (1914), 759. For European occurrences see the textbooks of Zirkel (*Petrography*), Tschermak, etc.

[3] Cf., for instance, the siliceous oölites of Pennsylvania, in Brown, *op. cit.*, pp. 760-68.